

# Tight-binding study of the electronic and magnetic properties of an $L1_0$ ordered FeCu alloy

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## Abstract

We have calculated the electronic structure of the tetragonal  $L1_0$  ordered FeCu by solving self-consistently a tight-binding Hamiltonian for s, p and d electrons. We have found by total energy calculation that this structure is ferromagnetic. In addition, we have determined that the equilibrium ratio between the interlayer and the intralayer lattice parameters is 0.947.

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Magnetic multilayers synthesized by ultrahigh vacuum deposition techniques have received much attention recently for both the fundamental and technological points of view[1, 2]. Particularly, the fabrication of ordered alloys with layered structure has created widespread interest because they may have important application potential in magnetic recording[3]. A typical example is the tetragonal  $L1_0$  ordered structure shown in Fig. 1, which consists of alternate atomic monolayers of two different elements. The magnetic properties of artificially fabricated Fe/Pt and Fe/Au multilayers with  $L1_0$  structure has been experimentally investigated[4]. Moreover, theoretical calculations of the magnetic and structural properties of Fe/Au monoatomic multilayers have been made by means of the FLAPW method[5]. All these systems have been found to be ferromagnetic (FM) and to have a large uniaxial magnetic anisotropy perpendicular to the atomic layers. Another system which promises interesting magnetic properties is the tetragonal  $L1_0$  ordered FeCu. The fabrication of this alloy has not been reported yet, but it seems possible since it is widely believed that fcc Fe ( $\gamma$ -Fe) films grow coherently on the Cu(100) surface due to the small lattice mismatch (1.1 %). In fact, layer-by-layer growth has been reported for Fe/Cu(100) films grown by thermal deposition[6] and has been greatly improved very recently by pulsed laser deposition[7]. In these systems, the layers of Fe are found to be individually FM, but regarding to the interlayer coupling, ferromagnetism has been observed only for the first layers, whereas the deeper ones exhibit antiferromagnetism[8, 9].

Motivated by these results, we have undertaken, for the first time to the best of our knowledge, a theoretical investigation of the electronic structure of the tetragonal  $L1_0$  ordered FeCu. Our calculations have been performed using the self-consistent tight-binding (TB) method within the unrestricted Hartree-Fock (HF) approximation by considering s,p and d orbitals. We have chosen this approach because it has shown to give succesful results in several magnetic systems as Fe clusters[10] and Fe monolayers sandwiched in noble metals[11]. In addition, this formalism has the advantage that it is posible to increase the complexity of the model by adding one by one different contributions to the Hamiltonian. Whitin this simple model, we have found, by total energy calculations, that the  $L1_0$  ordered FeCu alloy is FM with a magnetic moment of

2.69  $\mu_B$  and that the ratio of the interlayer lattice constant ( $c$ ) to the intralayer lattice constant ( $a$ ) is 0.947. As there are neither experimental data nor theoretical results available in the literature over the artificial Fe/Cu L1<sub>0</sub> structure, we limit ourselves to compare the present results with the related Fe/Au L1<sub>0</sub> structure.

The spin-polarized electron structure of the system is determined by solving self-consistently the TB Hamiltonian including hopping to second nearest neighbors. The Hamiltonian in the HF approximation has the general form:

$$H = \sum_{\vec{i}, m, \sigma} E_{\vec{i}m\sigma} n_{\vec{i}m\sigma} + \sum_{\vec{i} \neq \vec{j}, m, m', \sigma} T_{\vec{i}\vec{j}}^{mm'} a_{\vec{i}m\sigma}^\dagger a_{\vec{j}m'\sigma} \quad , \quad (1)$$

where  $a_{\vec{i}m\sigma}^\dagger$  ( $a_{\vec{i}m\sigma}$ ) and  $n_{\vec{i}m\sigma}$  are the creation (annihilation) and number operators of an electron state at atomic site  $\vec{i}$  in the orbital  $m$  with spin  $\sigma$ .  $E_{\vec{i}m\sigma}$  and  $T_{\vec{i}\vec{j}}^{mm'}$  stand for the single HF energies and the hopping integrals. The value of these integrals, for Fe-Fe and Cu-Cu interactions, are taken from Ref. [12] and for Fe-Cu interactions we employ the geometrical averages of the preceding ones.

The electron-electron interaction and the magnetic effects are introduced only in the diagonal terms of the Hamiltonian through the on-site potential shift of the energy levels,

$$\Delta E_{\vec{i}m\sigma} = \sum_{m'} U_{\vec{i}mm'} \Delta n_{\vec{i}m'} - \frac{\sigma}{2} J_{\vec{i}m} \mu_{\vec{i}m} \quad , \quad (2)$$

where  $U_{\vec{i}mm'}$  and  $J_{\vec{i}m}$  are the intra-atomic Coulomb and exchange integrals. To reduce the number of parameters we have assumed  $U_{\vec{i}ss} = U_{\vec{i}sp} = U_{\vec{i}pp}$  and  $U_{\vec{i}sd} = U_{\vec{i}pd}$ ; thus, we have only three kinds of intra-atomic Coulomb integrals. The value of these parameters for Fe and Cu were estimated as in the work by Sarma[13]. The exchange integrals were neglected with the exception of the one corresponding to Fe d electrons, whose value ( $J_{Fe} = 1.19$  eV) was chosen to recover the proper magnetic moment of bulk fcc Fe ( $\mu_{Fe} = 2.5 \mu_B$ ). Regarding to  $\Delta n_{\vec{i}m}$  and  $\mu_{\vec{i}m}$ ,  $\Delta n_{\vec{i}m} = n_{\vec{i}m} - n_{\vec{i}m}^0$  is the difference between the number of electrons determined by  $n_{\vec{i}m} = \langle n_{\vec{i}m\uparrow} \rangle + \langle n_{\vec{i}m\downarrow} \rangle$  and the occupation in the bulk metallic configuration  $n_{\vec{i}m}^0$ , and  $\mu_{\vec{i}m} = \langle n_{\vec{i}m\uparrow} \rangle - \langle n_{\vec{i}m\downarrow} \rangle$  is the local magnetic moment. Both magnitudes are determined from  $\langle n_{\vec{i}m\sigma} \rangle$ , which

is calculated self-consistently through:

$$\langle n_{i\sigma} \rangle = \int_{-\infty}^{E_F} \rho_{i\sigma}(E) \delta E \quad , \quad (3)$$

where the Fermi level ( $E_F$ ) is determined from the global charge neutrality condition, and the spin-polarized local density of states (SLDOS)  $\rho_{i\sigma}(E)$  is calculated by the recursion method[14]. The number of k levels of the continuous fraction expansion (k=30) is chosen so that  $\rho_{i\sigma}(E)$  becomes independent of k.

Systematically we have calculated the electron structure of the present system as a function of the volume. Intralayer ferromagnetism with both FM and antiferromagnetic (AFM) coupling between Fe layers were considered and  $a = a_{Cu} = 3.61 \text{ \AA}$  was assumed. By calculating the total energy, taking care of the double counting of the Coulomb and magnetic interactions, we determine the stability of the two different phases and the optimum ratio  $c/a$ . The results are plotted in Fig. 2. There it can be seen that the FM phase is more stable because is lower in energy than the AFM phase. By fitting the total energy through a Murnaghan equation of state[15], we determine for the FM case an equilibrium ratio  $c/a = 0.947$ , which is very close to the value estimated from the hard sphere radii of Cu and Fe ( $c/a = 0.974$ )[16]. This optimum ratio corresponds to a contraction of about 5 % with respect to the Cu volumen and results to be larger than the value obtained in total energy calculations in the Fe/Au L1<sub>0</sub> structure[5].

Next we focus on magnetization. For  $c/a = 0.947$  we have found that Fe has a magnetic moment ( $\mu_{Fe}$ ) of  $2.69 \mu_B$ . In addition, we have obtained that the atoms of Cu have a small magnetic moment ( $\mu_{Cu}$ ). Both moments are plotted as a function of  $c/a$  in Fig. 3. It can be seen there that  $\mu_{Fe}$  increases from  $2.49 \mu_B$  to  $2.81 \mu_B$  and  $\mu_{Cu}$  decreases from  $0.112 \mu_B$  to  $0.038 \mu_B$  as  $c/a$  variates from 0.8 to 1. This is just the behaviour that one would expect, because in general an enhancement of the magnetic moment occurs when diluting the Fe atoms and no magnetic moment would be expected in isolated Cu atoms.

To understand the enhancement of  $\mu_{Fe}$  and the origin of  $\mu_{Cu}$  it is a good help to give a look to the SLDOS. Fig. 4 shows the local density of states corresponding to the FM case for the equilibrium  $c/a$  value. The spin-up peaks around -2 eV and

spin-down peaks around 0 eV are mainly d electrons of Fe. The increase of the splitting between spin-up and spin-down peaks in comparison with the bulk splitting explains the enlargement of the Fe magnetic moment. Regarding to the peaks around -4 eV, they are related to d electrons of Cu atoms. It can be seen in Fig. 4 that they are a little bit shifted from each other giving rise to the small magnetic moment of Cu atoms. The existence of such small induced magnetic moment in Cu is related with the coupling between Fe monolayers and Cu monolayers.

Comparing again with the Fe/Au L1<sub>0</sub> structure, it results that in the present system  $\mu_{Fe}$  is less enhanced. We think that this difference may be due to two facts: the first one is that the indirect exchange interaction between Fe layers across noble metal layers decreases monotonically along the series Cu  $\rightarrow$  Ag  $\rightarrow$  Au [17]. The second one is that, d electrons of Cu are much closer in energy from those of Fe than d electrons of Au (situated around -5 eV[5]) and, as a consequence of the larger overlap of d electrons, the hybridization between Fe and Cu becomes larger than the hybridization between Fe and Au. This also allows us to understand why  $\mu_{Cu} > \mu_{Au}$ . Then, as we have obtained,  $\mu_{Fe}$  must be less enhanced in the Fe/Cu L1<sub>0</sub> structure than in the Fe/Au L1<sub>0</sub> structure because of the smaller degree of confinement experienced by Fe monolayers in the former system due to the larger indirect coupling and hybridization strength. We think that another possible consequence of this facts would be the decrease of the strong magnetic anisotropy observed in Fe/Au structures[4]. In any case, only comparison with already non-existent experimental work would allow us to check our predictions.

In summary, we have investigated theoretically the electronic and magnetic properties of the Fe/Cu monoatomic multilayer by means of a tight-binding scheme. We have found that in the tetragonal L1<sub>0</sub> structure the FM coupling between Fe layers is more favorable than the AFM one. For the FM case we determine that the ratio  $c/a$  has a value of 0.947 and the magnetic moment of Fe atoms is  $2.69 \mu_B$ . From our results, we conclude that Fe/Cu L1<sub>0</sub> structure belongs to the tetragonal L1<sub>0</sub> family of ferromagnets. Nevertheless, further progress is strongly needed for accurate understanding of its properties.

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## Figure captions

**Figure 1:** Crystal structure of the tetragonal  $L1_0$  ordered structure.

**Figure 2:** Relative total energy as a function of the volume for the tetragonal  $L1_0$  ordered FeCu. Circles are the calculated values when ferromagnetic ( $\bullet$ ) or antiferromagnetic ( $\circ$ ) coupling is considered. Lines are least square fits to the Murnaghan equation of state.  $V_0$  is the equilibrium volume of fcc Cu.

**Figure 3:** Calculated magnetic moment of Fe ( $\bullet$ ) and Cu ( $\circ$ ) in the tetragonal  $L1_0$  ordered FeCu as a function of  $c/a$ . Lines are only a guide to the eye.

**Figure 4:** Local density of states (SLDOS) for the tetragonal  $L1_0$  ordered FeCu. The vertical solid line represents the Fermi level.









